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ATOMIC HYDROGEN DISSOCIATORS FOR LOW TEMPERATURE  
ATOMIC HYDROGEN MASERS  
AND  
IN-VACUUM DISSOCIATORS FOR VLG-11 SERIES MASERS  
GRANT NAG-8012  
FINAL REPORT



For the period from 20 April 1981 through 26 April 1984

Principal Investigator  
Dr. Robert F.C. Vessot

(NASA-CR-173506) ATOMIC HYDROGEN FOR LOW  
TEMPERATURE ATOMIC HYDROGEN MASERS AND  
IN-VACUUM DISSOCIATORS FOR VLG-11 SERIES  
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Astrophysical Observatory  
Cambridge, Massachusetts 02138

The Smithsonian Astrophysical Observatory  
is a member of the  
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The NASA Technical Officer for this grant is Dr. Rudolph Decher,  
Space Physics Division, Code ES61, Marshall Space Flight Center, Alabama 35812

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ATOMIC HYDROGEN DISSOCIATORS FOR LOW TEMPERATURE

ATOMIC HYDROGEN MASERS

AND

IN-VACUUM DISSOCIATORS FOR VLG-11 SERIES MASERS

GRANT NAG-8012

Two types of R and D activities were funded by NASA's Marshall Space Flight Center (MSFC) on this grant:

1. Develop an r.f. dissociator which can be installed within the vacuum envelope of a cryogenic hydrogen maser.
2. Design, build an in-vacuum dissociator for the VLG-11 series maser and test it in a VLG-10 maser.

The first activity was funded by MSFC on April 20, 1981 and the second activity was funded by MSFC on September 14, 1982.

1.0 DISSOCIATORS FOR LOW TEMPERATURE ATOMIC HYDROGEN MASERS

1.1 INTRODUCTION

Hydrogen masers require a source of hydrogen atoms in the form of a directed beam. In conventional masers operating at room temperature the source of atoms is nearly in thermal equilibrium with the storage region. In the cold masers, however, a conventional atomic hydrogen source would be much hotter than the storage volume. This can cause at least two types of problems. First, there is radiative heat transfer from the source to the cold cavity and second, there is the question of thermal equilibrium of the hot, fast-moving hydrogen atoms after they enter the cold cavity and the effect they can have dislodging the wall coating atoms frozen to the surface. An atomic hydrogen source cooled

at least to the liquid nitrogen temperature helps to diminish these problems.

The object of this project is to study the operation of a cryogenically-cooled hydrogen maser using an r.f. plasma dissociator operating at liquid nitrogen temperature (77K) in conjunction with a state selector magnet whose dimensions are suitable for slow atoms. The focussing characteristics for a hexapole state selector magnet with maximum fields at the pole tips,  $H_m$ , provide a maximum acceptance angle for atoms at the most probable velocity in the beam given by

$$\theta_m = \left[ \frac{2\mu_0 H_m}{3kT} \right]^{\frac{1}{2}}$$

where  $\mu_0$  is the Bohr magneton and  $kT$  is the thermal energy per unit bandwidth,  $k$  is Boltzmann's constant, and  $T$  is the absolute temperature of the beam source.

Since the acceptance solid angle is proportional to  $\theta_m^2$ , the magnet is approximately 4 times more effective at 77K than at 300K. Furthermore, (though this is not a very important consideration) the magnet's length is reduced by about one-half.

By thermally isolating the r.f. circuitry from the dissociator glassware, only dielectric losses in the glass and the energy coupled to the plasma will result in the boil-off of liquid nitrogen. We estimate that this is about one watt and thus anticipate a loss rate of approximately .022 liters per hour. This rate can easily be accommodated in the presently available experimental setup.

## 1.2 THE EXPERIMENTAL VACUUM ENCLOSED R.F. DISSOCIATOR

During the past three years, S.A.O. has had an ongoing r.f. hydrogen dissociator program supported by NASA in which different types of dissociator enclosures are being life tested under various conditions of r.f. excitation and cooling. Several conclusions have so far been drawn.

1. The glasses that seem to work best are borosilicate glasses, Corning 7070 and 7740.
2. The dimensions of the dissociator bulb should be on the order of 4 cm; smaller tubes seem prone to excessive surface recombination.
3. The glass can run fairly hot, 50-60°C without any obvious loss of performance.
4. Because of the non-critical cooling requirements, a dissociator can be operated within the vacuum enclosure rather than being in the atmosphere. Conductive cooling of the glass to a heat conducting "sink" is all that is required. This means that for operation in the vacuum of space there is no need for an active cooling system as used in the 1976 Redshift space probe maser. It also means that difficult and costly glass-to-metal vacuum seals are not required; simple ground joints will suffice since hydrogen leakage from the low pressure (<1 Torr) dissociator to the vacuum system is negligible when compared to the high beam flux from the collimator.

The basic design of the dissociator which was used in the test system was particularly simple. It was later used in the four experimental and advanced development models of XDM and ADM passive hydrogen masers, developed and tested for the U.S. Naval Research Laboratory. As with the other systems, these operated from an external r.f. supply at 80-100 MHz, capable of delivering about 5 watts to loads of highly variable impedance resulting from the fact that the hydrogen presents very different electrical characteristics to the generator when it is ionized in a plasma state.

This type of dissociator, with the bulb outside of the vacuum enclosure, was originally adopted for the cold maser, as shown in Figure 1. However, it soon became obvious to us that we were headed for trouble with this structure. First, it was very difficult to maintain the alignment of the dissociator, which was attached to the outer vacuum enclosure, with the internal cryostat structure. Second, the heat leakage to the inside system was not insignificant. We therefore decided to equip the maser with the cold dissociator before making any further maser measurements at low temperatures.

The glass is thermally connected to a liquid nitrogen reservoir and enclosed in the vacuum system. A hexapole state selector magnet and beam stopping disc with dimensions suitable for the cryogenically cooled maser are attached to the cold structure. (See Figure 1a).

Figure 2 shows the device as it is built into the cryogenic maser. The liquid nitrogen cooled attachment ring is bolted to the cooled copper shroud as shown in Figure 3.



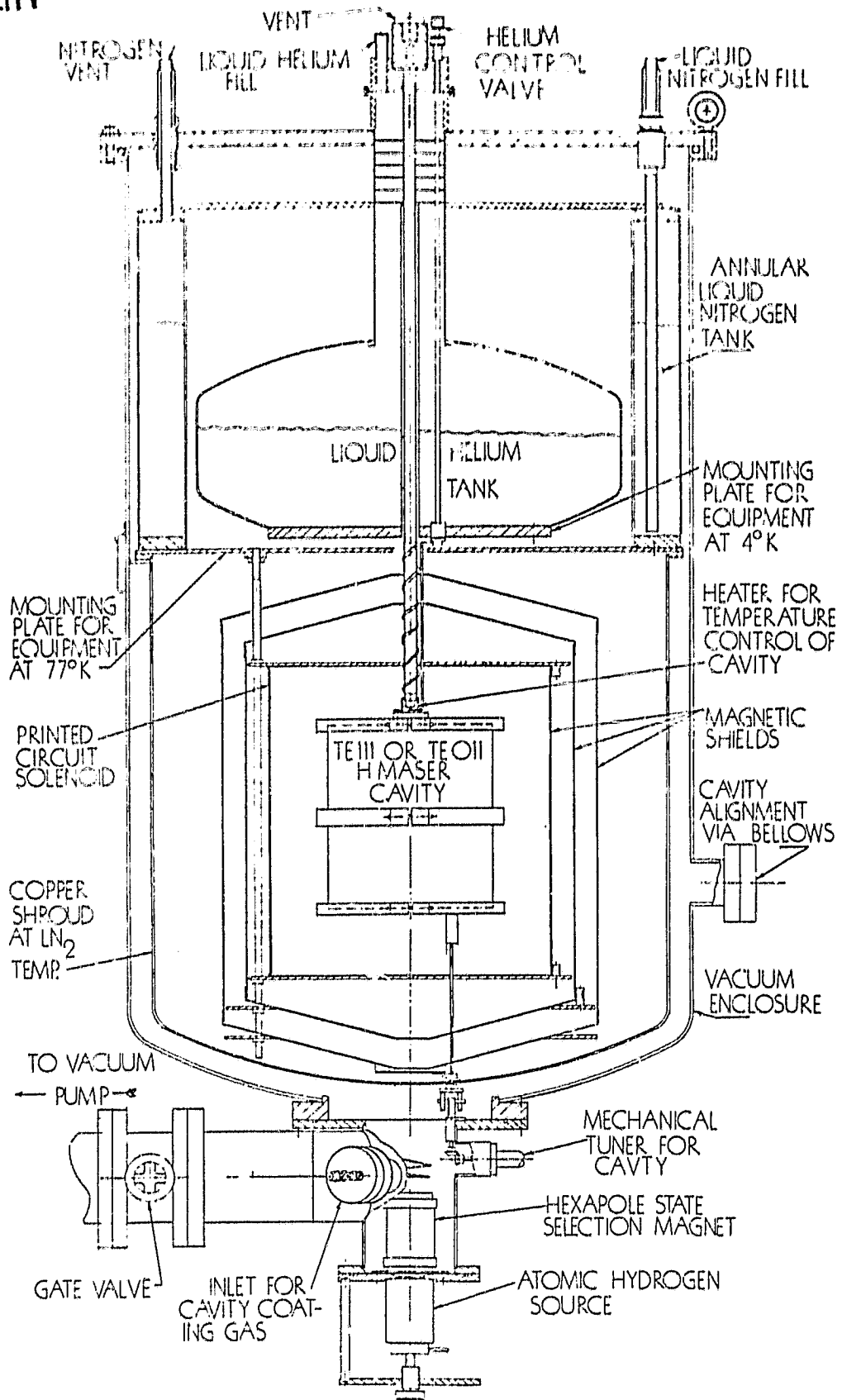


FIGURE 1 DESIGN OF CRYOGENIC MASER WITH ROOM TEMPERATURE DISSOCIATOR

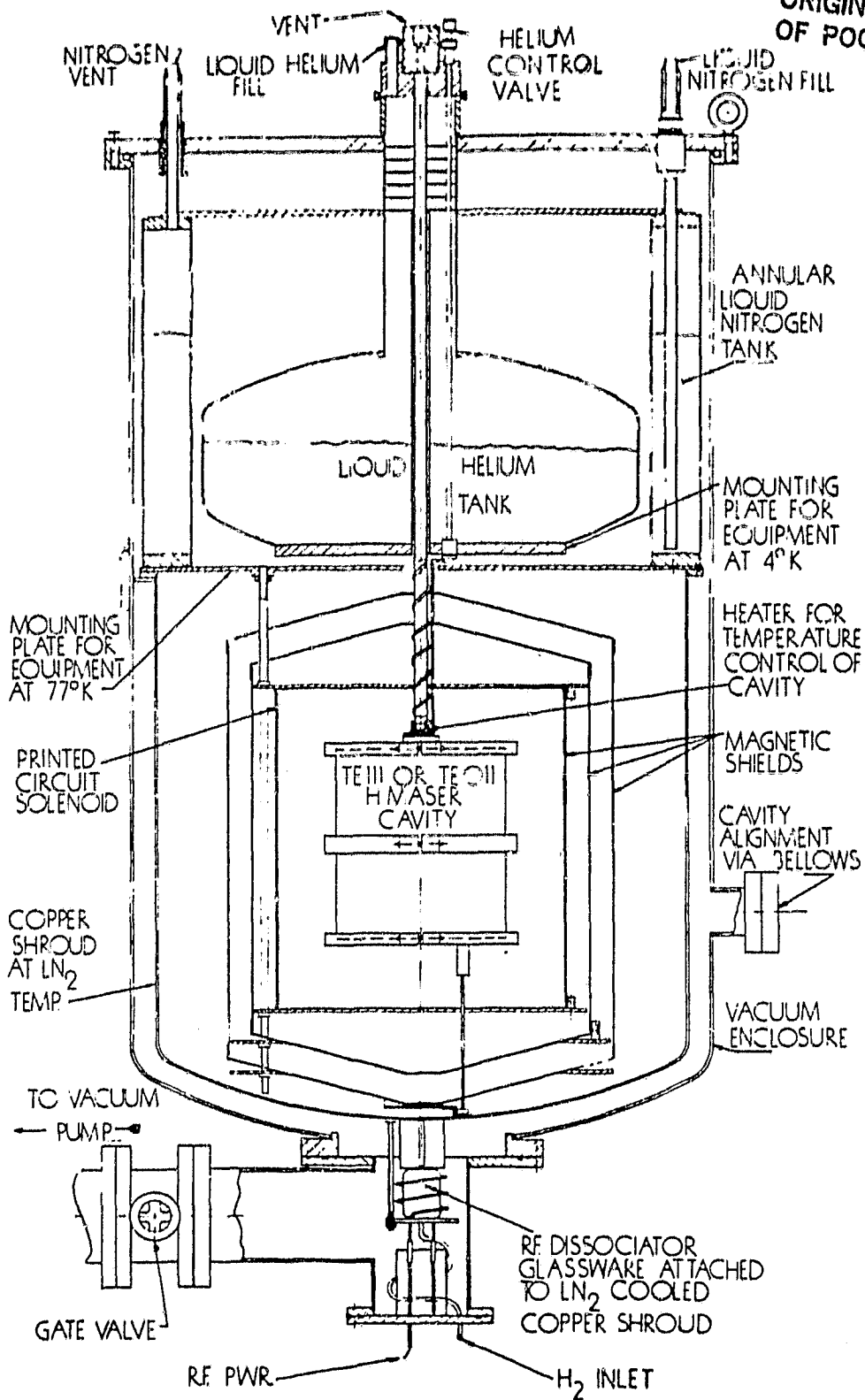


FIG. 1a SAO COLD MASER ASS'Y  
WITH COLD RF DISSOCIATOR

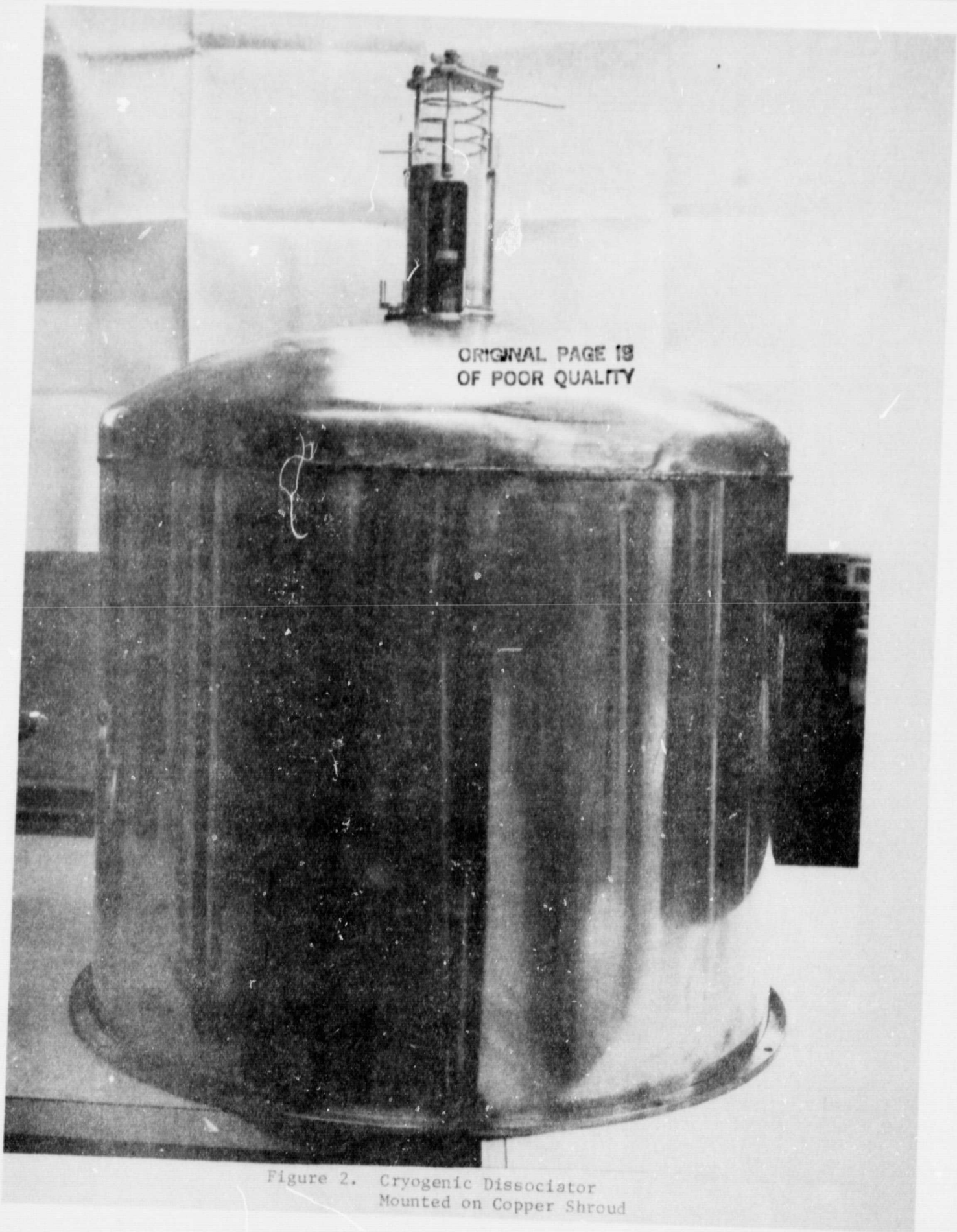


Figure 2. Cryogenic Dissociator  
Mounted on Copper Shroud

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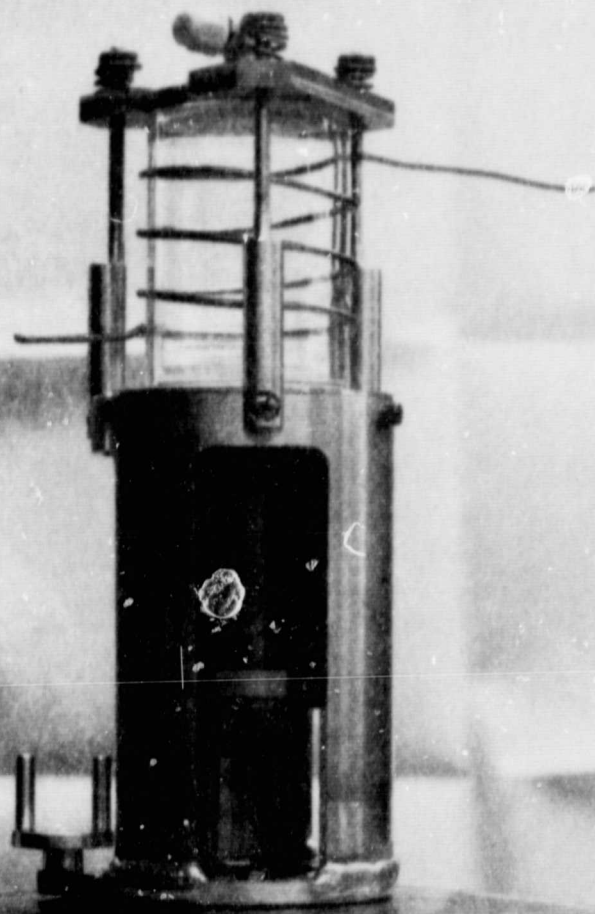


Figure 3. Closeup Showing LN  
Attachment Ring Bolted  
to Copper Shroud

### 1.3 STATUS OF THE PROJECT

By the end of April 1982, the dissociator as modified to operate in the vacuum of the cryogenic maser had been assembled and tested in vacuum at room temperature. During May and June 1982, the cryogenic maser was assembled and checked out. In late June, the (inner) helium tank was filled with liquid nitrogen, when cooled the hydrogen was turned on and flowing. The dissociator was lighted and the r.f. discharge looked good. (The r.f. source was at 57 MHz; r.f. generator power, 10 watts.)

At the end of June 1982, the inner and outer tanks of the dewar were filled with liquid helium and nitrogen respectively, the dissociator was lit briefly (r.f. source 44 MHz, 6 watts power) and the color was normal.

In mid-July, a cryogenic run of the cold maser was attempted. The dewar tanks were filled with liquid nitrogen. Although the dissociator functioned properly (59 MHz, 15 watt power), cold maser oscillation was not achieved. Ringing of the maser was achieved for a short period of time only at very high fields, but oscillation was not achieved. The problem suspected is bad magnetic conditions, possibly due to thermally-induced currents in the coax line to the cavity or in the magnetic field set up by the diode monitors.

The cryogenic maser cavity was reworked in the Fall of 1982, to shorten the cavity length to achieve oscillation at 77K. This work was done and the cryomaser reassembled in October-November, 1982. The tanks were refilled with liquid He and N<sub>2</sub> and maser oscillation was achieved. At this juncture, the funding for the cryogenic maser dissociator was expended. The dissociator has functioned flawlessly and made possible the results discussed in Appendix I.

## 1.4 CONCLUSION

We have demonstrated that the r.f. dissociator as modified to work in the vacuum of the cryogenic maser is a feasible design and that the dissociator operates satisfactorily at 77K. Work is continuing on cryogenic maser research until June 30, 1984 with funding from the Office of Naval Research. This research is directed towards measuring wall shift and relaxation rates of various wall coatings (i.e. Neon, fluorine ( $CF_4$ )) and attempt to achieve maser oscillation down to 4 Kelvins.

## 2.0 IN-VACUUM DISSOCIATOR FOR VLG-SERIES MASERS

### 2.1 HISTORY AND STATUS OF PROJECT

In early September 1982, M.S.F.C. authorized work on the in-vacuum dissociator for the VLG-11-series maser. Design effort commenced in September and the design and detail drawings were completed by October. The drawings were released to our Model Shop for fabrication of parts in late October. Fabrication of the dissociator parts was completed by late November 1982.

By mid-January 1983, the vacuum dissociator was assembled in a vacuum test station with a lithium aluminum hydride hydrogen source. The dissociator r.f. structure was matched to a frequency generator at 62.5 MHz and the r.f. amplifier was operated at 4.4 watts output power for the test. The dissociator operated at these levels for 4 weeks (until mid-February) and the r.f. discharge looked very good. At this point, the turbo-molecular pump failed and this test had to be discontinued.

In September 1982, SAO requested permission from NRL to use NRL's S/N F-0 maser as a test vehicle for the in-vacuum dissociator. (This maser is on loan

to SAO to perform maser R and D work for NRL, i.e. test a non-evaporable gettering system and perform cavity frequency stabilization system tests, etc.).

Permission was granted by NRL to use P-0 for the in-vacuum dissociator tests. Problems with P-0's bulb coating and cavity length delayed assembly of the in-vacuum dissociator until mid-June 1983, when the maser was reassembled with the new dissociator and pumped down. The maser was turned on, tuned and had the line Q measured. The maser oscillated satisfactorily, but at the end of June a gradual deterioration of the IF output level was observed. The maser stopped oscillating. On 5 July, the hydrogen pressure set was increased and the maser restarted. After 2 or 3 days of good oscillation, the IF slowly decreased and the maser stopped oscillating. The problem was traced to the Teflon solution used to coat the bulb in April 1983. This solution had deteriorated during storage at SAO, resulting in a poor bulb coating. A new Teflon solution was purchased in July and the bulb was recoated with the new solution in August. By mid-August 1983, the maser was reassembled, pumped down and turned on. The maser oscillated satisfactorily and the maser was tuned using USNO maser S/N P-18 as a reference.

By the end of September 1983, the in-vacuum dissociator had been operational for 1-1/2 months and was working very well. The r.f. output from the amplifier was 8 watts at 60.7 MHz. A characteristic of the vacuum dissociator is that the discharge appears dim in comparison to the VLG-11 masers which have their r.f. dissociator in air.

By the end of December 1983, (when the period of performance for this contract ran out) the in-vacuum dissociator had been in operation continuously and stably for 4-1/2 months. The testing has shown that the design is feasible and satisfactory and can be suitably adapted to the VLG series of hydrogen

masers. Although this unit has operated satisfactorily for over one-third of a year, further life testing is desirable to determine the long-term operating characteristics of the in-vacuum dissociator.

The design of the in-vacuum dissociator is illustrated by Figures 4 through 6. Figure 4 illustrates the method of mounting the dissociator glassware and the hexapole magnet to the vacuum flange. Figure 5 shows the outside face of the vacuum flange with the center viewing port, the 2 ports for the hydrogen system and the port for the r.f. input power for the r.f. coil on the dissociator glassware. Figure 6 shows the heat shield around the hexapole magnet.

In late January 1984, the maser started operating erratically and on 25 January, stopped oscillating -- the r.f. dissociator was off.

Subsequent testing showed we had r.f. heating problems inside. When the r.f. is tuned around 62 MHz, a sharp rise was noted in the ion pump current, indicating an r.f. short-circuit or onset of a glow discharge in the vacuum system. The test maser was opened and we saw a deposit of what appeared to be copper on the inside of the glass. This may have resulted from the reduction of copper oxide that got into the glass during assembly. The dissociator glassware was removed and both inside and outside surfaces were cleaned. In addition, the heat shield (around the hexapole magnet) was extended to protect the dissociator glassware from Corona discharge from the ion pump elements when they are started.

By the end of January 1984, the maser was reassembled and the Sorbac cartridges were activated. The maser was returned to the clock room and restarted. PO maser continued oscillating during March and has continued beating with SAO Maser S/N P-13. Although P-0 maser's frequency became occasionally erratic in late March, the cause is believed to be in the diode voltage control



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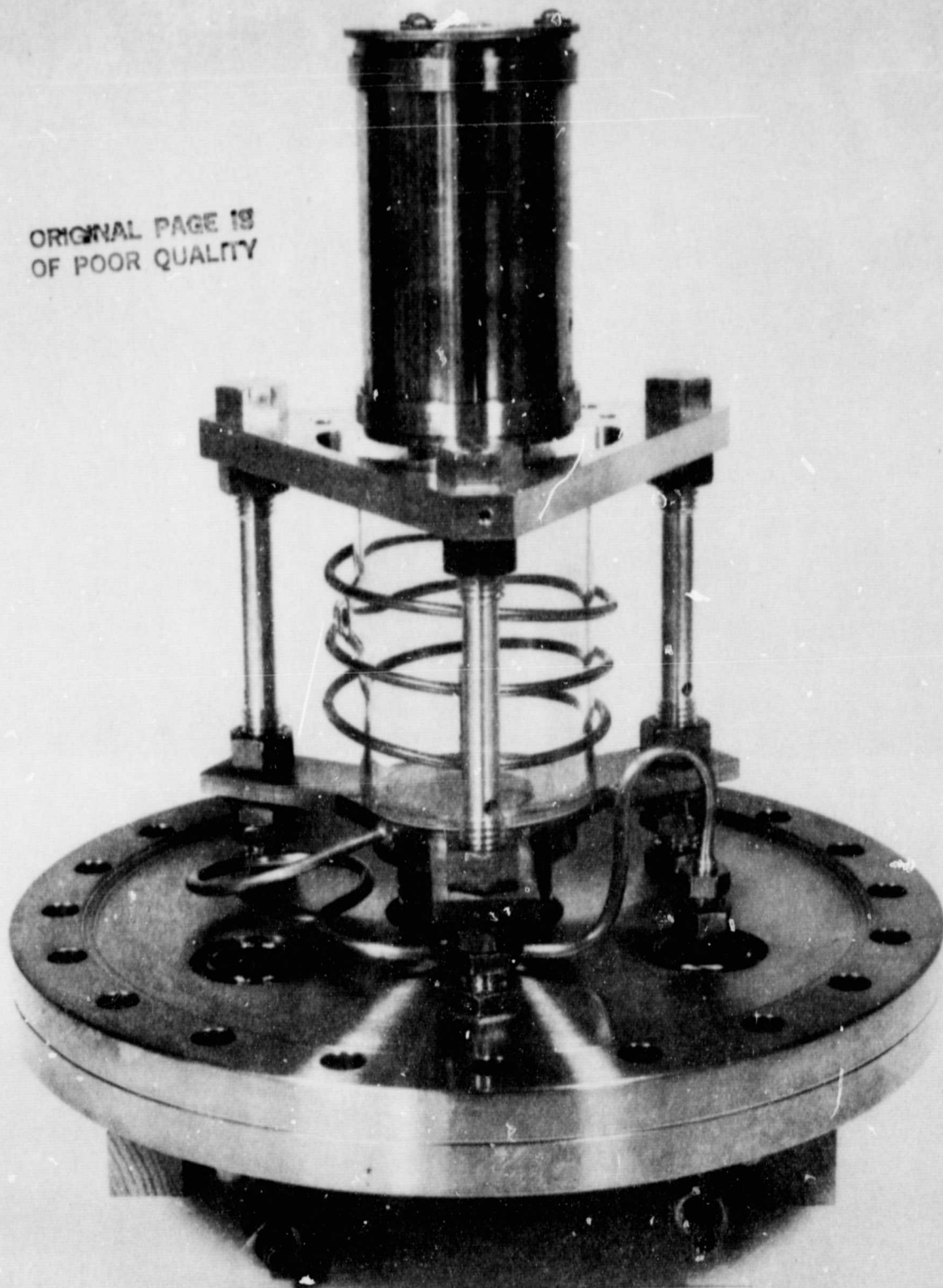


Figure 4. Method of Mounting  
Dissociator Glassware  
and Hexapole Magnet  
to Vacuum Flange

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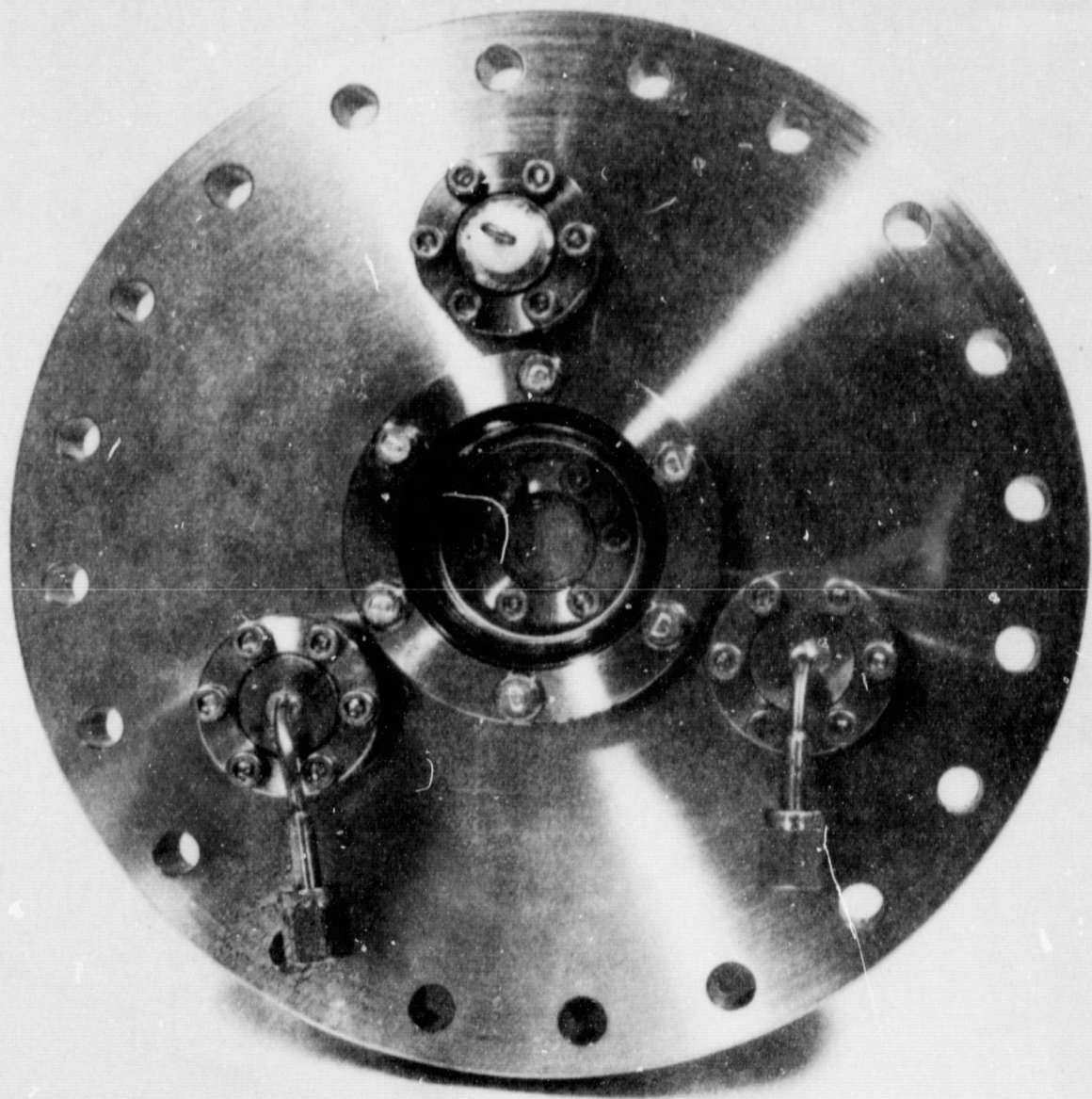


Figure 5. Outside Face of Vacuum  
Flange

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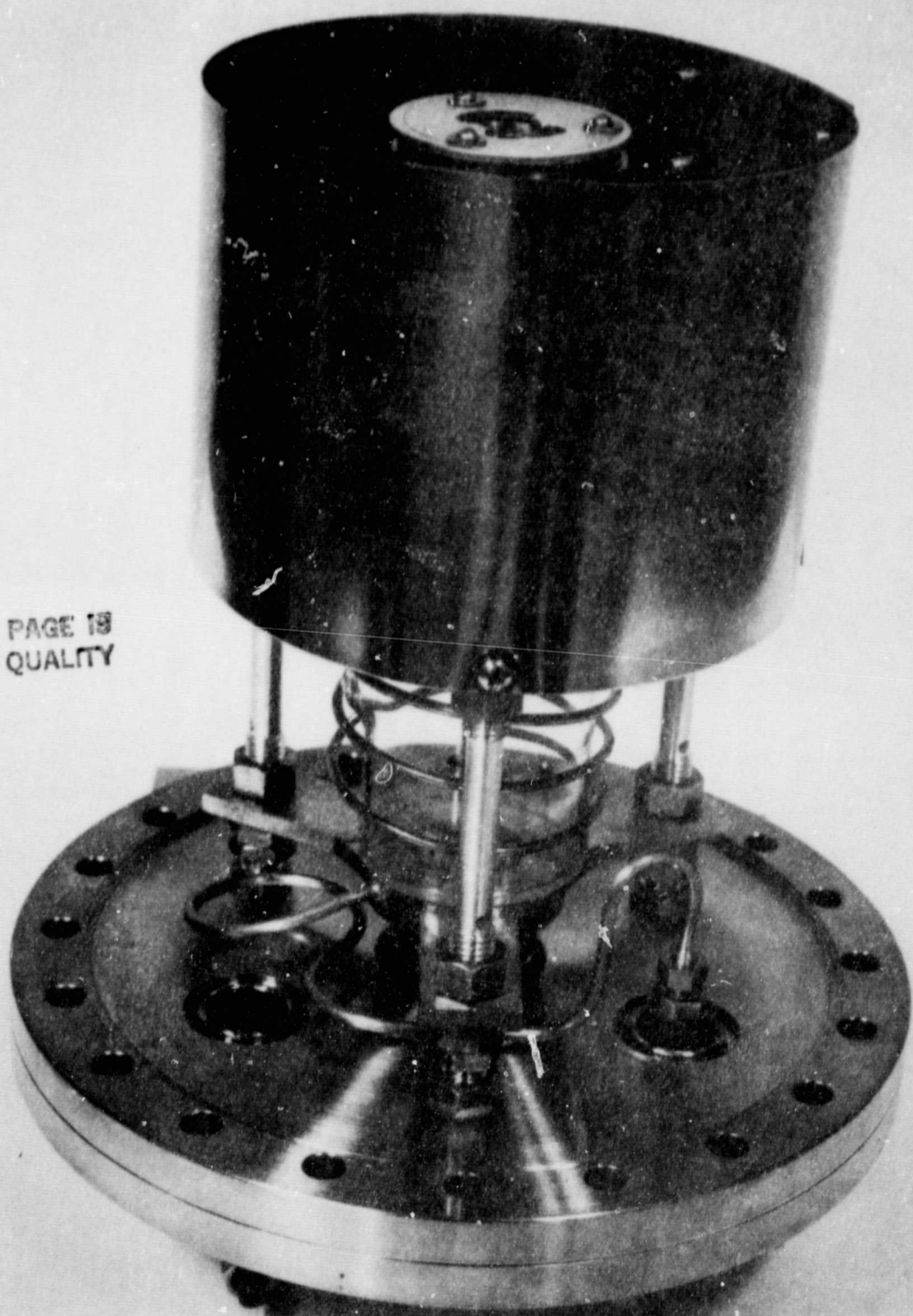


Figure 6. Heat Shield around  
Hexapole Magnet

circuitry and will be investigated. The in-vacuum dissociator has continued to operate satisfactorily through the end of this report period (26 April 1984).

## 2.2 CONCLUSION

Eight and one-half months of operation (mid-August 1983 to 26 April 1984) of the in-vacuum dissociator used in P-0 maser have shown that the design is viable and operational. Further life testing is required to see if there are any other long-term operating problems similar to the one discovered in January, 1984 where the glassware was contaminated by the Corona discharge of the ion pump elements. Also, the method of providing r.f. power for the dissociator, which can be integrated into the maser cabinet, is another area where further investigation is desirable.

APPENDIX I

PROCEEDINGS OF THE 37th ANNUAL SYMPOSIUM ON FREQUENCY CONTROL

1-3 JUNE 1983

pp 49-54

COLD HYDROGEN MASER RESEARCH AT SAO AND RELATED DEVELOPMENTS

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ABSTRACT

The cryogenically-cooled maser provides a means for measuring frequency shifts and relaxation properties of storage wall coating materials that can be frozen in place from substances normally in the gaseous phase. We report initial measurements for FEP-120 Teflon between 77K and 48K and compare them with the 372K to 77K data of de Saintfusien.<sup>1</sup> Some of the design features of the maser are described. The cryomaser's hydrogen dissociator is located entirely within the vacuum enclosure and operates at 77K. The in-vacuum dissociator and other developments that have been adapted and tested in a room temperature maser, and that are applicable for both space and terrestrial use, are also discussed.

INTRODUCTION

A program for studying the operation of atomic hydrogen masers at low temperatures has been under way at the Smithsonian Astrophysical Observatory since 1977. The aim of this program is to advance both the science and the technology of hydrogen masers. The work has been supported by the Office of Naval Research, the Smithsonian Institution, the National Aeronautics and Space Administration (NASA), the Jet Propulsion Laboratory, and Marshall Space Flight Center.

The principal reason for the outstanding stability of the hydrogen maser is the fact that hydrogen atoms can be confined in a storage vessel for extended periods (on the order of a second) with very little effect on the phase of their oscillating magnetic dipole moments. This moment results from the magnetic interaction between the proton and electron spins in the hydrogen ground state; the energy separation produced by this interaction provides the hydrogen maser signal, the familiar 21 cm line of radio astronomy.

Atoms stored in a hydrogen maser make many collisions with the wall surface of the storage volume. In conventional masers this surface is usually coated with Teflon, a long chain fluorocarbon. The interaction during collisions has not been extensively studied, and we are still using the same materials and techniques of 20 years ago. At present the theoretically available stability of the maser has been achieved for averaging periods between one second and one hour; in this regime the stability is limited by thermal noise, thermal movement of the atoms in the storage volume, and the rate of wall collision relaxation. This last process is what we want to investigate at low temperatures where electronic noise,  $kT$ , is reduced, and where atoms move more slowly and collide with much less energy than at room temperature.

By injecting substances that are gaseous at room temperature into the storage volume of a cryogenically cooled maser and allowing them to freeze on its walls, we can repeatedly coat the walls with different materials in a well-controlled manner. A measurement of the average advance or retardation of the phase of

the hydrogen atom's oscillating magnetic dipole (per collision) is obtained by measuring the frequency shift of the maser signal and knowing the kinetics of the atoms in the bulb. The average phase decorrelation is determined from the oscillation line Q, defined as

$$Q = \frac{\omega}{2\gamma_{2T}}$$

where  $\omega = 2\pi f$  is the maser frequency, and  $\gamma_{2T}$  is the relaxation rate of the phase from all causes, including exit of the atoms from the bulb. The line Q is analogous to the quality factor of any conventional oscillator and appears in the following expression for the fundamental limitation of frequency stability:

$$\sigma_{fy}(\tau) = \frac{1}{Q} \left[ \frac{kT}{2P_b \tau} \right]^{\frac{1}{2}}$$

This is the one-sigma expectation of the fractional frequency variation measured in the time interval  $\tau$ . Here  $kT$  is the thermal noise power per unit bandwidth ( $k$  is Boltzmann's constant,  $T$  is absolute temperature) and  $P_b$  is the power generated by the oscillator, in this case the state-selected hydrogen atoms that have been focussed into the storage volume.

Additive white phase noise also contributes to the frequency instability; the variance of the fractional frequency variations from this source is given by

$$\sigma_{ay}(\tau) = \frac{1}{2\pi f \tau} \left[ \frac{FkTB}{P_o} \right]^{\frac{1}{2}}$$

where  $F$  is the receiver noise figure,  $B$  is the overall system bandwidth in Hz, and  $P_o$  is the power delivered to the receiver.

There are several reasons to expect increased hydrogen maser stability at low temperatures.

1.  $kT$  is smaller. This also affects the signal-to-noise ratio of equipment that receives the maser signal.
2.  $P_b$  can be made larger. The limit on  $P_b$  is from hydrogen-hydrogen spin exchange collisions in the storage volume. The cross section for such collision diminishes rapidly with temperature making it possible to have a much greater density for a given level of spin exchange quenching.
3. The lower speed of the atoms reduces the wall collision rate and, all other things being equal, correspondingly increases the storage time.
4. Many properties of materials are much better behaved at low temperatures and excellent magnetic field control can be accomplished using superconducting shields.

OBJECTIVES OF THE COLD MASER RESEARCH PROGRAM

The principal objective of our present efforts is to determine the behavior of the various types of walls as a function of temperature and to develop an understanding of the fundamental processes governing the collision interactions.



The possible advantages of operating a hydrogen maser at temperatures well below room temperature have been speculated upon for many years, but until recently the prospects for successfully operating such a device at temperatures significantly below 77K seemed marginal. Kleppner and his co-workers<sup>2</sup> demonstrated that atomic hydrogen could be contained at 4K in a vessel whose walls were coated with frozen molecular hydrogen. This gave considerable impetus to investigate the behavior of the hyperfine resonance at temperatures below 77K, which was the temperature limit explored by M. de Saintfusien and others<sup>1</sup> in their research into wall relaxation and wall coatings.

In 1977 work began at SAO on an initial version of a liquid helium-cooled cryostat that contained a TE<sub>111</sub>-mode split storage volume resonator with fluorinated ethylene propylene copolymer (FEP) Teflon coated walls. At first, oscillation could only be sustained down to temperatures of about 55K. Why it would not oscillate below this temperature was open to many questions -- were the walls of the storage volume contaminated because of the rather crude vacuum system we employed, or was there a more fundamental limit imposed by the nature of the atomic hydrogen in its collision with the FEP surfaces? To answer these questions we decided to try a different approach with the cold maser. This was to introduce carbon tetrafluoride (CF<sub>4</sub>) gas through the hydrogen source structure and freeze a coating of this molecule on the inside surfaces of the cavity storage volume. This technique provided a fluorine-bonded-to-carbon surface comparable to the long chain fluoride molecules of Teflon. The scheme worked, and greatly to our delight, we were able to maintain oscillation down to temperatures of 26K.

This work was reported<sup>3</sup> at the 33rd Annual Symposium on Frequency Control and we continued theoretical studies to determine what advantages would accrue if we could operate at or below 26K.<sup>4</sup> From theory we know that the stability would be well below 1 part in 10<sup>16</sup> at 1000 seconds provided no new instability problem arose. However, we know that the large temperature-dependence of the wall shift is likely to cause difficult requirements for temperature stabilization in the cold maser.

#### MECHANICAL CONFIGURATION OF THE COLD MASER

The design of the maser cryostat required considerable effort and imagination to anticipate as many uses as possible to which it could be put in a cold maser testing program. First, it had to be structurally reliable. Second, it had to have a reasonably large volume in order to house a variety of possible maser cavity and magnetic shield configurations. We decided that if we had uncertainties in these dimensions it would be better to have a larger, rather than smaller, volume. As an upper limit, we decided that it should be able to house the full size TE<sub>011</sub>-mode cavity and inner magnetic shield design such as was used in the lightweight space-probe maser.<sup>5</sup>

The question of the range of operating temperatures was also an issue. Our previous success in obtaining oscillation at temperatures as low as 26K suggested that we should cover the range from 77K to somewhere near 4.2K with provision for stabilization at any point between these temperatures. This was done by mounting the maser cavity on a tubular support that contains a constant-loss, metered flow of liquid helium that cools the cavity and offsets heat leakage into the system. By metering a slightly greater flow of helium than required to offset the heat leak, an electrical resistance heater on the cavity support can

be used in a servo-controlled system to hold the cavity temperature at the desired level.

For temperatures below the liquid helium boiling point, we included provisions to change our method of cooling and can connect the cavity directly to the helium bath. By pumping on the helium gas, we should be able to drive the temperature well below 4K.

Additionally, we wanted to have a reasonably large capacity for helium so that disturbances from filling could be avoided for up to five days of steady operation. Since the projected cavity and shielding structures that would be cooled tended to be quite massive and the apparatus itself would be rather large, we decided to use a liquid nitrogen guard region to surround the liquid helium container and the sample region. This reduces the cost of cryogenics used for initial cooling and allows more flexibility of operation. It also eliminates the need for multi-layer reflective insulation (superinsulation) and the ensuing outgassing problems that could lead to surface contamination in the atomic hydrogen storage volume of the maser.

The final design of the SAO cold hydrogen maser cryostat is shown in Figure 1. The fact that the liquid helium and nitrogen tanks are above the region to be cooled is dictated by the need for a gravity feed of cryogen to the cavity region.

The annular 30 liter liquid nitrogen tank is thermally connected to an equipment mounting plate of copper beneath it and to a heavy copper shroud surrounding the working volume, which is roughly 22 inches (56 cm) in diameter x 22 inches (56 cm) deep. Inside it is the 30 liter liquid helium tank, which has a flat base plate for mounting equipment to operate near 4K. The cavity resonator is suspended from a tubular sample holder that is thermally isolated from the liquid helium reservoir by being attached to a point near the top of the dewar, allowing it to operate at temperatures substantially above 4K. Liquid helium flows to the sample holder through a needle valve that controls its flow rate. The magnetic shield assembly is suspended by thermally conducting rods from the nitrogen tank and is kept near 77K to prevent the excessive loss of magnetic shielding owing to the reduction of magnetic permeability at very low temperatures.

All the components are mounted on the 26 inch (66 cm) diameter top plate of the cryostat which is vacuum sealed by an O-ring to the vacuum enclosure. Access is gained to the assembly by lifting the plate out of the enclosure using a crane as shown in Figure 2.

To obtain reasonably fast pumping, as well as good enough cleanliness to permit outgassing the system at the at the 10<sup>-8</sup> torr level, we use a turbomolecular pumping system with 450 liter/second throughput. This is intended to pump the system when it is at or above 77K. When liquid helium is used, the 4 inch diameter gate valves are closed and the system cryopumps, so that all gases but helium condense on the helium tank.

The cavity resonator used in the first tests was a 15 cm diameter x 18 cm long TE<sub>111</sub>-mode r.f. cavity<sup>6</sup> (instead of the usual 28 cm diameter x 28 cm diameter long TE<sub>111</sub>-mode cavity). This cavity is equipped with a Teflon septum to separate the two regions of r.f. magnetic field that are 180 degrees out of phase. The remaining interior surfaces of the cavity are coated with FEP-120 (fluorinated ethylene propylene copolymer). The entry collimator is split by the sep-

tum, each side leading to one-half the cavity,

The gas handling system for the wall coating gases utilizes a ballast volume, metering valves, and pressure- and flow-measuring equipment. A mechanical scavenging pump allows control of the gas ballast tank pressure, as well as purging and cleaning of the system. The gas is admitted through a valve and led to two jets located on the downstream side of the hexapole magnet, each aimed into one of the input collimators of the TE<sub>111</sub> cavity's storage volume.

#### THE COLD HYDROGEN DISSOCIATOR

Hydrogen masers require a source of hydrogen atoms in the form of a directed beam. In conventional masers operating at room temperature the source of atoms is nearly in thermal equilibrium with the storage region. In the cold masers, however, a conventional atomic hydrogen source would be much hotter than the storage volume. This can cause at least two types of problems. First, there is radiative heat transfer from the source to the cold cavity and second, there uncertainty in the method of thermal equilibration of the hot, fast-moving hydrogen atoms after they enter the cold cavity and the possibility of their dislodging the wall coating atoms frozen to the surface. A beam source cooled at least to liquid nitrogen temperature would substantially help to diminish these problems.

We have designed a cryogenically-cooled hydrogen maser using an r.f. plasma dissociator operating at liquid nitrogen temperatures (77K) in conjunction with a state selector magnet whose dimensions are suitable for slow atoms. The focusing characteristics for a hexapole state selector magnet with maximum fields at the pole tips,  $H_m$ , provide a maximum acceptable angle  $\theta_m$  for atoms at the most probable velocity in the beam given by

$$\theta_m = \left[ \frac{2\mu_0 H_m}{3kT} \right]^{1/2}$$

where  $\mu_0$  is the Bohr magneton.

Since the acceptance solid angle is proportional to  $\theta_m^2$ , the magnet is approximately 4 times more effective at 77K than at 300K. Furthermore -- although this is not a very important consideration -- the magnet's length is reduced by about one-half.

By thermally isolating the r.f. circuitry from the dissociator glassware, only dielectric losses in the glass and the r.f. energy coupled to the plasma will result in the boil-off of liquid nitrogen. We estimate that this is about one watt and thus anticipate a loss rate of approximately .022 litres per hour.

The design of the dissociator is particularly simple and was later used in the four experimental- and advanced-development models of passive hydrogen masers, developed and tested by SAO for the U.S. Naval Research Laboratory.<sup>7</sup> As with the other systems, these operated from an r.f. oscillator and power amplifier at 80-100 MHz, and are capable of delivering about 5 watts to the dissociator. The load impedance presented by the dissociator is highly variable because the hydrogen presents very different electrical characteristics to the generator when it is ionized in a plasma state than when it is a neutral gas.

The glass of the dissociator bulb is thermally connected to the liquid nitrogen reservoir and is enclosed in the vacuum system. A hexapole state selector magnet and beam stopping disc with dimensions

suitable for the cryogenically cooled maser are attached to the cold structure.

Figure 3 shows the device as it is built into the cryogenic maser. The liquid nitrogen cooled attachment ring is bolted to the cooled copper shroud as shown in Figure 1.

The very low power dissipated by the glass and the plasma within it has led us to try this type of design in non-cryogenically cooled masers. Here the heat from the glass is conducted to the vacuum envelope, and the excitation coil, made of heavy copper, is also thermally connected to the vacuum envelope.

This design, where the dissociator is wholly within the vacuum envelope, eliminates the need for elastomer seals and is particularly attractive for use in spaceborne masers where the thermal control of the dissociator is best done by conductive means rather than by a recirculated air flow system as used in the 1976 Redshift space maser.<sup>5</sup>

#### RESULTS OF WALL COATING MEASUREMENTS AS OF MAY 1983

The two measurable wall properties are:

1. The wall collision frequency shift. This is expressed in terms of the average phase shift per collision.
2. The wall relaxation rate. This is expressed in terms of the probability of loss of phase per collision.

Our present efforts have been to measure the wall frequency shift. The temperature dependence of the phase shift per collision gives some important clues about the nature of the interaction between impinging hydrogen atom and the wall surface. The perturbation results from polarization of the hydrogen's electron wave function owing to a dipole-dipole attractive force as the atom approaches the wall and a strong, repulsive interaction at short ranges arising from Pauli exclusion exchange forces. Two types of frequency shifting effects are encountered: 1) The polarization of the hydrogen atom leads to negative frequency shifts because of the diminished interaction of the displaced electron cloud with the proton, and 2) Pauli forces lead to positive frequency shifts resulting from compression of the electron cloud about the hydrogen atom and stronger interaction.

These two can offset and cancel each other. The temperature at which this occurs depends on the surface properties<sup>8-11</sup>. At low temperatures the negative shift dominates and can be expressed as an exponential function of the temperature,  $e^{E/kT}$ . Here  $E/k$  can be considered an energy expressed in degrees Kelvin.

Our data are shown in Figure 4, where we plot the logarithm of the average phase shift per collision against inverse temperature. From 77K to approximately 52K the data follow the relationship:

$$\Delta\theta = 6.17 \times 10^{-6} e^{215/T}$$

As had occurred in the earlier experiments,<sup>3</sup> during the cooling and measuring process the maser stopped oscillating at about 50K. Whether this was due to contamination or was a consequence of the nature of the REP Teflon surfaces is still not clear. At this point we admitted gaseous CF<sub>4</sub> into the system, raising the pressure to 10<sup>-4</sup> Torr for 30 seconds, and waited for the surplus to pump away. The next day we took data, then allowed the system to warm up to about 60K and took more data as the system cooled again. These data are shown as open circles in Figure 4 and were



obtained at temperatures down to 48K. These data have a different relationship with temperature than that of the bare FEP Teflon surface:

$$\Delta\theta = 11.6 \times 10^{-6} \cdot 165/T$$

From this run we observe that the re-cooled data do not fit the first data point obtained after injecting  $CF_4$ . It is possible that the surfaces were not heavily coated with  $CF_4$  and that reevaporation (or resublimation) occurred when we went to 60K, leaving some of the Teflon surface exposed.

During this run we suffered a blockage in the helium flow and were unable to cool lower than 48K. However, in our earlier cryostat we were able to get oscillation at temperatures as low as 26K, but were unable to run at low enough magnetic fields to make accurate wall shift measurements.

The data reported in Figure 4 agree well with those of de Saintfuscion.<sup>1</sup> Figure 3 shows his data and ours plotted on the same graph. Here we see the effects of phase changes in the Teflon that occur near 300K and 200K. A summary of the properties of FEP Teflon as they would apply for a maser with a 7 inch (17.8 cm) diameter bulb is given in Table 1. We show the wallshift, its temperature coefficient, and the quantity  $(df/f)/(dT/T)$ , which measures the fractional temperature sensitivity. In general, at low temperatures the fractional temperature can be controlled at least as well as at higher temperatures. Nevertheless, we see that to realize frequency stability of  $1 \times 10^{-16}$  will require temperature control at the  $5 \times 10^{-6}K$  level at 50K, not a simple matter.

### CONCLUSIONS

It is clear that we are looking for a surface material that does not alter the chemical state of the impinging hydrogen atom. Beyond this, we require as low an interaction potential with hydrogen as possible so that the atom is reflected without dwelling too long in a potential well.

So far we are aware of only a small number of solid and liquid materials that have been investigated as storage surfaces. The wallshift interaction energies and their applicable temperatures are shown in Table 2 for some interesting examples. Of the Teflons, we include here only the FEP fluorocarbon. Surfaces of liquid helium<sup>4</sup> and helium<sup>3</sup> have been measured at very low temperatures by Hardy. These are extremely interesting surfaces with very low interaction energies and should lead to some interesting results when applied to hydrogen masers.<sup>12,13</sup>

So far the fluorocarbon surfaces appear to work as low as 25K. If we consider what the fluorine atom looks like when bound to carbon, we note that its electronic structure is that of the noble gas neon, which has the next closed electron shell occurring after helium. It would appear that fluorine, when it looks like neon to an outsider, works well. It is logical, then, to try neon itself as the surface coating material. This is where we are headed as soon as we can get our system to function at temperatures between 8K and 14K.

The expected frequency stability of cold masers, based on thermodynamics and spin exchange behavior, is well below  $1:10^{-16}$  for averaging times of 100 seconds.<sup>4,13</sup> The potential for greatly increased stability is a strong incentive for continuing the study of wall interactions.

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Table 1

Performance of Maser with  
7-inch (17.8 cm) Diameter Bulb

Temp (K)	Surface Coating	Wallshift $\Delta f/f$	Wallshift Temperature Coefficient $(df/dT)/f$  ( $K^{-1}$ )	$(df/f)$ $(dT/T)$
323	FEP	$-1.03 \times 10^{-11}$	$2.21 \times 10^{-13}$	$7.1 \times 10^{-11}$
150	FEP	$-3.76 \times 10^{-11}$	$3.14 \times 10^{-13}$	$4.7 \times 10^{-11}$
100	FEP	$-7.38 \times 10^{-11}$	$1.57 \times 10^{-13}$	$1.5 \times 10^{-10}$
60	FEP	$-2.37 \times 10^{-10}$	$1.20 \times 10^{-11}$	$7.2 \times 10^{-10}$
50	FEP + CF <sub>4</sub>	$-3.07 \times 10^{-10}$	$1.72 \times 10^{-11}$	$8.6 \times 10^{-10}$

Table 2

Interaction Energies and Temperatures of  
Various Surfaces with Atomic Hydrogen

Surface	Reference	E/K (K)	T (K)
FEP Teflon	1	458	300
FEP Teflon	this work	263	77
FEP Teflon	this work	215	-50
FEP + CF <sub>4</sub>	this work	165	-48
Neon	—	$\sim 400$	10-20(?)
Solid H <sub>2</sub>	2	38	4.2
Liquid He <sub>4</sub>	12	1.15	-0.1
Liquid He <sub>3</sub>	12	0.43	<0.1

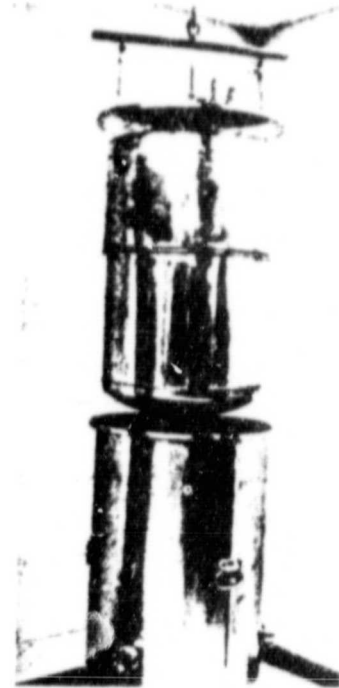


Fig. 2. Cryogenic maser suspended from vacuum housing.

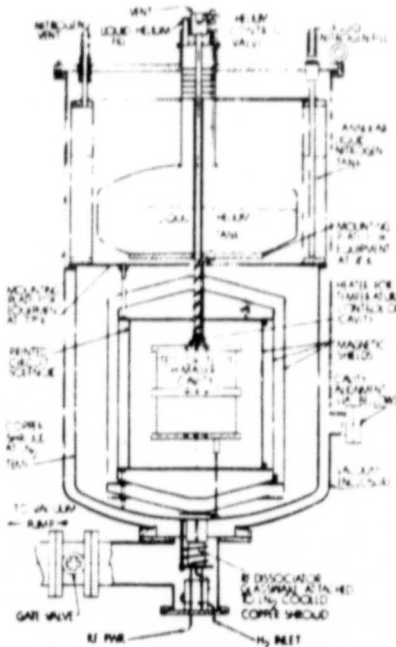


Fig. 1. Cryogenically cooled hydrogen maser with low temperature dissociator.

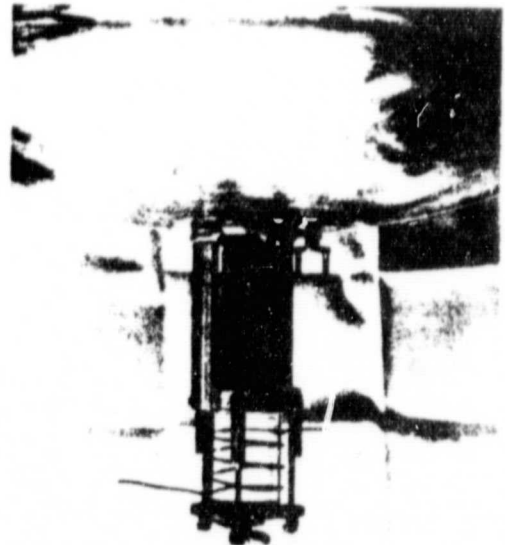


Fig. 3. Liquid nitrogen cooled hydrogen dissociator.

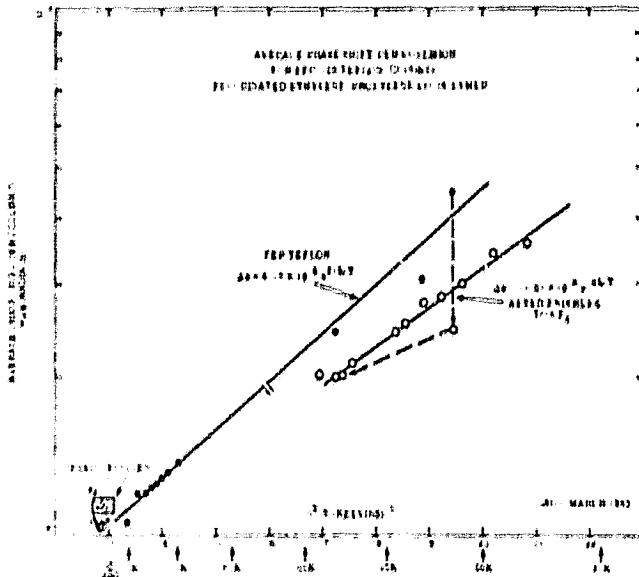


Fig. 4. Average phase shift per collision,  $-\Delta\theta$  (radians), from 77K to 48K.

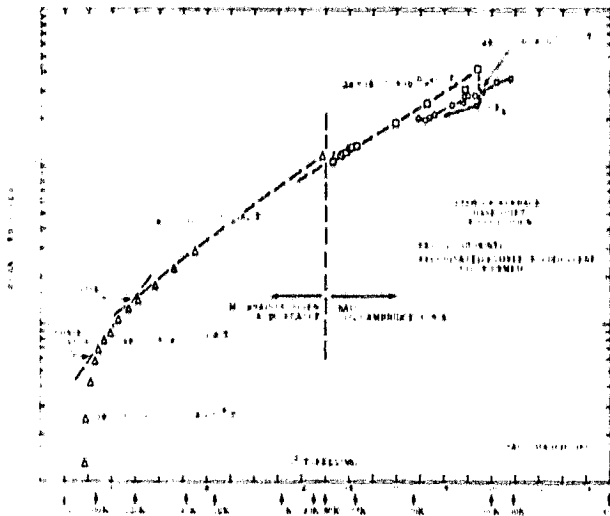


Fig. 5. Average phase shift per collision,  $-\Delta\theta$  (radians), from 330K to 48K.